Brominated Flame Retardants in North-East Atlantic Marine Ecosystems

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Background: Concentrations of brominated flame retardants (BFRs) are reported to increase in marine ecosystems.

Objectives: Characterize exposure to BFRs in animals from different trophic levels in North-East Atlantic coastal marine ecosystems along a latitudinal gradient from southern Norway to Spitsbergen, Svalbard, in the Arctic. Calanoid species were collected from the Oslofjord (59°N), Froan (64°N), and Spitsbergen (> 78°N); Atlantic cod (*Gadus morhua*) from the Oslofjord and Froan; polar cod (*Boreogadus saida*) from Bear Island (74°N) and Spitsbergen; harbor seal (*Phoca vitulina*) from the Oslofjord, Froan, and Spitsbergen; and ringed seal (*Phoca vitulina*) from Spitsbergen. Eggs of common tern (*Sterna hirundo*) were collected from the Oslofjord, and eggs of arctic terns (*Sterna paradisaea*) from Froan and Spitsbergen.

Results: Levels of polybrominated diphenylethers (PBDEs) and hexabromocyclododecane (HBCD) generally decreased as a function of increasing latitude, reflecting distance from release sources. The clear latitudinal decrease in levels of BFRs was not pronounced in the two tern species, most likely because they are exposed during migration. The decabrominated compound BDE-209 was detected in animals from all three ecosystems, and the highest levels were found in arctic tern eggs from Spitsbergen. HBCD was found in animals from all trophic levels, except for in calanoids at Froan and Spitsbergen.

Conclusions: Even though the levels of PBDEs and HBCD are generally low in North-East Atlantic coastal marine ecosystems, there are concerns about the relatively high presence of BDE-209 and HBCD.

Key words: Arctic, biomagnification, HBCD, hexabromocyclododecane, Norway, PBDE, polar bear, *Ursus maritimus*, polybrominated diphenylethers, seals. *Environ Health Perspect* 115(suppl 1):35–41 (2007). doi:10.1289/ehp.9355 available via *http://dx.doi.org/* [Online 8 June 2007]

Brominated flame retardants (BFRs) are technical flame retardants containing brominated organic compounds which are applied to combustible materials, such as plastics, wood, paper, and textiles to meet fire safety regulations (Alaee et al. 2003; de Wit 2002). Additive flame retardants, such as polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCD), are blended with the polymers and may leach out of the products (Alaee et al. 2003). Being environmentally persistent compounds with high production volumes, PBDEs and HBCD are among the most abundant BFRs detected in the environment (Alaee et al. 2003).

The predominant commercial PBDE products are penta-, octa- and deca-BDE technical mixtures, and these have been produced and used in large volumes (Darnerud 2003). The penta-BDE (Bromkal 70) consists of mainly tetra- and pentabrominated congeners (i.e., BDE-47 and BDE-99, respectively), whereas the octa-BDE contains mainly hepta-, octa-, and nonabrominated diphenylethers [e.g., BDE-183 (hepta) and BDE-203 (octa)]. The use of the penta-BDE and octa-BDE mixtures is prohibited in all applications for the European Union Market since August 2004 (European Union 2003). Deca-BDE, consisting mostly of BDE-209, is believed to be less threatening to the environment because its large molecular size is assumed to limit its global atmospheric transport potential and its bioavailability (de Boer et al. 2003; De Wit 2002; Gouin et al. 2006). There are currently no restrictions on the use of technical deca-BDE products (de Boer et al. 2003), and the same is the case for HBCD (Stapleton et al. 2006).

BFRs are lipophilic and many are resistant to physical and biochemical degradation. Such BFRs are therefore bioaccumulative and may biomagnify in food webs, and are thus classified as persistent organic pollutants (POPs). Concentrations of PBDEs in marine mammals, birds, and fish have been increasing in recent decades, although the concentrations found in the European wildlife are in general lower than those found in North America (Hites 2004). Temporal studies have also documented that levels of HBCD are increasing in seals in North America (Stapleton et al. 2006) and in harbor porpoises (*Phocoena phocoena*) in the United Kingdom (Law et al. 2006b).

Recently, there has been particular focus on the dispersal and bioaccumulation of the highly brominated PBDEs, such as BDE-209. This congener is believed to bioaccumulate less in marine ecosystems than in terrestrial ecosystems. However, recent reports of BDE-209 in Glaucous gulls (*Larus hyperboreus*) and polar bears (*Ursus maritimus*) (Sørmo et al. 2006; Verreault et al. 2005) indicate that this congener has properties that allow it to disperse into the Arctic and to bioaccumulate in homeothermic marine predators. Furthermore, relatively high levels of HBCD have been reported in homeothermic marine animals (Law et al. 2006b; Morris et al. 2004; Murvoll et al. 2006a; Zegers et al. 2005), even in the Arctic (Murvoll et al. 2006b; Sørmo et al. 2006). Thus there are particular needs for identifying the bioaccumulation and biomagnification potential of BDE-209 and HBCD in marine ecosystems.

Many BFRs have detrimental effects on organisms (Darnerud 2003; De Wit 2002). Identification of spatial trends in exposure, bioaccumulation, and biomagnification of these compounds in marine ecosystems is important in risk assessment of BFRs for wildlife health.

The aim of the present study was to characterize exposure to seven PBDE congeners [BDE-28 (2,4,4′-tribromodiphenyl ether), BDE-47 (2,2′,4,4′-tetrabromodiphenyl ether), BDE-99 (2,2′,4,4′,5 -pentabromodiphenyl ether), BDE-100 (2,2′,4,4′,6 -pentabromodiphenyl ether), BDE-153 (2,2′,4,4′,5,5′ -hexabromodiphenyl ether), BDE-154 (2,2′,4,4′,5′,6 -hexabromodiphenyl ether), BDE-209 (2,2′,3,3′,4,4′,5,5′,6,6′-decabromodiphenyl ether)] and HBCD in animals from different trophic levels in North-East Atlantic coastal marine ecosystems along a latitudinal gradient from southern Norway to Spitsbergen, Svalbard, in the Arctic (Figure 1). Samples were collected from the southern

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Oslofjord (59°N), Froan on the west coast of Norway (64°N), Bear Island in the Barents Sea (74°N), and Spitsbergen (> 78°N). Calanoid species were obtained from the Oslofjord, Froan, and Spitsbergen; Atlantic cod (*Gadus morhua*) from the Oslofjord and Froan; polar cod (*Boreogadus saida*) from Bear Island and Spitsbergen; harbor seal (*Phoca vitulina*) from the Oslofjord, Froan, and Spitsbergen; and ringed seal (*Phoca vitulina*) from Spitsbergen. Eggs of common tern (*Sterna hirundo*) were collected from the Oslofjord, and eggs of arctic terns (*Sterna paradisaea*) were collected from Froan and Spitsbergen.

Material and Methods

Animals. Organisms were sampled in the southern Oslofjord (Hvaler: 59° N, 11° E), at Froan in the Norwegian Sea on the west coast of Norway (64° N, 9° E), at Bear Island in the Barents Sea (74° 27′ N, 19° E), and at Spitsbergen (> 78° N, 10–15° E). The calanoid species were collected using zooplankton (1,000 µm mesh) trawls at depths of 0–350 m. Polar cod (Sørmo et al. 2006) and Atlantic cod were caught using trawls or fishing rods. The sample sizes (*n*) of Atlantic cod were 20 in the Oslofjord and at Froan, respectively. The sample sizes of polar cod from Bear Island and Spitsbergen were 6 and 7, respectively. In the Oslofjord (Ruus et al. 2002) and at Froan, blubber samples were collected from healthy adult male harbor seals that had been shot (*n* = 5 and 9, respectively). At Spitsbergen (Prins Karls Forland, 79° 54′ N, 10° 36′ E), blubber biopsies were obtained from healthy live-caught adult male harbor seals (*n* = 5), and blubber samples were collected from 5 healthy adult male ringed seals (Forlandssundet, 78° 20′ N) that had been shot (Sørmo et al. 2006). Eggs of common terns were collected at Hvaler (*n* = 10), whereas eggs of arctic terns were collected at Froan (*n* = 10) and at Spitsbergen (Hotellneset, Longyearbyen, 78° 14′ N, 15° 29′ E) (*n* = 10) during the early egg-laying periods. When possible, biometric data on the animals were taken. All samples were collected during 2003, except for the blubber samples of harbor seals from the Oslofjord which were collected in 1998 (Ruus et al. 2002).

Culling of seals and collection of blubber biopsies and eggs were approved by Norwegian authorities (Ruus et al. 2002; Sørmo et al. 2006), and animals were treated humanely and with regard for alleviation of suffering.

Samples of zooplankton were wrapped in aluminium foil and stored in 50-mL polyethylene containers. Individual specimen samples of whole polar cod and blubber/adipose tissue samples from the seals were wrapped in aluminium foil and stored in plastic bags. All samples were kept frozen at –20°C.

We estimated the biomagnification potentials of the BFR compounds assuming a simple cod–harbor seal food chain model. The biomagnification factor (BMF) for each of the compounds was calculated as $BMF_X = [C_{Xpred}]/[C_{Xprey}]$, where BMF_X is the biomagnification factor of compound X, $[C_{Xpred}]$ is the mean concentration of compound X in the predator, and $[C_{Xprey}]$ is the mean concentration of compound X in the prey. BMFs were estimated using lipid weight concentrations of the compounds.

The data presented on levels of BFRs in *Calanus glacialis*, polar cod, and ringed seals from Spitsbergen have been presented elsewhere (Sørmo et al. 2006), but are included herein to elucidate spatial distributions in BFRs in marine ecosystems in the North-East Atlantic. Details on the sampling procedures in the Oslofjord and at Spitsbergen are given elsewhere (Ruus et al. 2002; Sørmo et al. 2006).

Analytical methods for BFRs. The chemical analyses of BFRs were conducted at the Laboratory of Environmental Toxicology at the Norwegian School of Veterinary Science in Oslo using gas chromatography-mass spectrometry (GC-MS) analysis. Pooled samples (~150 g) of the calanoid species from each of the locations (*n* = 1–3, Table 1) were crudely homogenized in a food blender. Whole Atlantic and polar cod (~ 10 g), blubber from harbor and ringed seals (~ 2 g), and whole eggs of common and arctic terns (~ 20 g) were homogenized separately with scalpels in Petri dishes. The homogenates were transferred to 80-mL centrifuge tubes, and an internal standard mix (100 ng/mL) of BDE-77, BDE-119, BDE-181, and ^{13}C-BDE-209 (Cambridge Isotope Laboratories, Inc., Andover, MA, USA) was added to each sample. A detailed description of the extraction procedure and of the methods for separation and detection of PBDEs (including BDE-209) and HBCD is given by Sørmo et al. (2006). An aliquot of 1 mL from all samples was evaporated to dry condition on a sand bath (ST7; H. Gestigkeit GmbH, Düsseldorf, Germany) at 40°C for gravimetrical determination of the extractable lipid content.

In all GC-MS analyses, the temperature quadropole was set to 106°C, ion source to 250°C, and interface to 300°C. The GC-MS was operated in the electron capture mode with methane (Hydro Gas, Oslo, Norway) of purity 4.7 as reagent gas. To monitor the different BFRs, we used selected ion monitoring. The PBDEs (except BDE-209) were monitored at *m/z* 79/81. HBCD was monitored at *m/z* 79/81 and 159.8. BDE-209 was monitored at *m/z* 484 and 486 and ^{13}C-BDE-209 at *m/z* 495 and 497. Electron energy of 86.6 eV was used (Sørmo et al. 2006).

Chromatographic data were calculated using the software MSD ChemStation G1701 version C.00.00 (Agilent Technologies, Santa Clara, CA, USA). Concentrations of the individual BFRs were determined by corresponding components in the standards, and analyzed for BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-209, and HBCD. Quality assurance for the analyses included a 6- to 8-point linear calibration curve of the analyzed standard solutions. Detection limits were set to about 3 times the noise level and varied among species and chemicals: 0.012–1.299 ng/g lipid weight (lw) in invertebrates, 0.030–0.30 ng/g lw in Atlantic and polar cod, and 0.014–0.75 ng/g lw in the harbor and ringed seal blubber. HBCD consists of the three diastereomers α-, β-, and γ-HBCD. At temperatures > 160°C in the injection port, as used in this GC analysis, thermal rearrangement of the diastereomers leads to isomeric interconversion of β- and γ-HBCD to α-HBCD (Peled et al. 1995); thus, our results predict total HBCD.

We used internal standards to detect and correct changes in compound concentrations during the chemical preparation and injection of the extracts into the GC-MS run. We also analyzed recovery of samples of corn oil spiked with BFR standard solutions after each sample series. Mean percent recovery and coefficient of variance of the individual BFRs in the corn oil samples ranged from 70 to 115% and from 1 to 28%, respectively. Standard solutions were run every 10 samples during the GC-MS analysis to detect any drift in the responses of

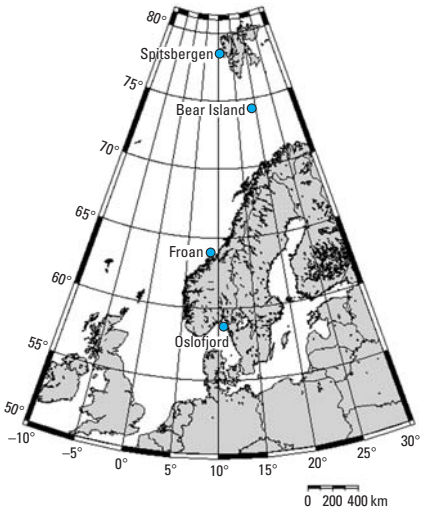

Figure 1. Calanoids were collected in the Oslofjord (59° N), at Froan (64° N), and at Spitsbergen, Svalbard (> 78° N). Atlantic cod (*Gadus morhua*) were collected in the Oslofjord and at Froan, polar cod (*Boreogadus saida*) at Bear Island (74° N) and Spitsbergen, harbor seals (*Phoca vitulina*) in the Oslofjord, at Froan, and at Spitsbergen, and ringed seals (*Phoca hispida*) were collected at Spitsbergen. Eggs of common terns (*Sterna hirundo*) were collected in the Oslofjord, whereas eggs of arctic terns (*Sterna paradisaea*) were collected from Froan and Spitsbergen. See text and Sørmo et al. (2006) for more specific sampling locations at Spitsbergen.

the analysis. We tested reproducibility over time continuously by analyzing the laboratory's own control (seal blubber) at a minimum of one sample per series. Concentrations of the components in the seal blubber control were compared with the mean of previous years; they were within 2 times the standard deviation of the mean. The laboratory is accredited by Norwegian Accreditation (Kjeller, Norway) for testing BFRs in biological material of animal origin according to the requirements of the Norwegian Standard–English Standard International Organization for Standardization/International Electrochemical Commission (NS–EN ISO/IEC 17025, Test 051). The laboratory's analytical quality for BFRs (not including BDE-209 and HBCD) was approved in intercalibration tests (de Boer and Wells 2004). Because of high levels of BDE-153 in the blanks of the polar cod batch, coinciding with high levels of this compound in polar cod extracts, this compound was not validated and reported in polar cod (Sørmo et al. 2006).

Concentrations of the BFR compounds are presented on a lipid-weight basis in homogenates of calanoid species, in liver and whole body homogenates of individual Atlantic and polar cod, in blubber of harbor and ringed seals, and in eggs of common and arctic terns.

Statistical analyses. Statistical analyses were conducted using SPSS version 11.5 (SPSS Inc., Chicago, IL, USA). For *Calanus glacialis*, the results are represented by one observation. Because of the relatively small samples sizes, with undetermined distributions, we used nonparametric Kruskal-Wallis (when comparing more than two groups/species) and Mann-Whitney tests (when comparing two groups/species) to compare BFR concentrations among the locations and the different trophic levels. Significance was set at $p < 0.05$.

Results

Calanoids. In the Oslofjord, all BFR compounds were detected in the sampled calanoid species (Table 1). BDE-47 was the most abundant compound, followed by BDE-99. In *Calanus finmarchicus* from Froan, only BDE-47, BDE-99, and BDE-100 were detected, and BDE-47 was the most abundant compound. In *Calanus glacialis* from Spitsbergen, only BDE-47 and -99 were detected, and the concentrations of these congeners were similar. Thus, ΣPBDEs in calanoids from the Oslofjord, Froan, and Spitsbergen, represented 7, 3, and 2 BDE congeners, respectively, and ΣPBDE was higher in calanoids from the Oslofjord than from Froan (ΣPBDEs: Mann-Whitney *U*-test, $n = 6$, $z = -1.964$, $p = 0.05$) and from Spitsbergen (not tested because $n = 1$ at Spitsbergen) (Table 1, Figure 2A). The levels of BDE-47 and BDE-99, which were found in calanoid species from all three locations, also decreased as a function of increasing latitude (Table 1). HBCD was detected in calanoids only from the Oslofjord (Figure 3A).

Atlantic and polar cod. All BFR compounds were detected in the Atlantic cod from the Oslofjord and Froan (Table 1). BDE-47, followed by HBCD, was the most abundant compound in Atlantic cod from both these locations. Levels of ΣPBDEs (Figure 2B; Mann-Whitney *U*-test, $n = 42$, $z = -3.324$, $p < 0.001$), but not HBCD (Figure 3B; Mann-Whitney *U*-test $n = 39$, $z = -1.916$, $p = 0.057$), differed between Atlantic cod from the Oslofjord and Froan.

In polar cod from Bear Island, five of the nine compounds (BDE-28, BDE-47, BDE-100, BDE-154, HBCD) were detected, whereas in polar cod from Spitsbergen, seven of the compounds were detected (BDE-28, BDE-47, BDE-99, BDE-100, BDE-154, BDE-209, HBCD) (Table 1). At both locations, HBCD was the most abundant compound, followed by BDE-47. In polar cod from Spitsbergen, relatively high concentrations of BDE-209 were found in five of the seven specimens. Concentrations of ΣPBDEs (Figure 2B) and HBCD (Figure 3B) were significantly higher in polar cod from Bear Island compared with those from Spitsbergen (Mann-Whitney *U*-test, $n = 13$, $z = -3.000$, $p = 0.001$ for both ΣPBDEs and HBCD).

Levels of ΣPBDEs and HBCD differed between the four locations [Kruskal-Wallis, $\chi^2 > 26.63$, degrees of freedom (df) = 3, $p < 0.001$]. When comparing levels of ΣPBDEs and HBCD in the two cod species from all four locations, levels of ΣPBDEs were Oslofjord > Froan > Bear Island > Spitsbergen (Figure 2B), whereas levels of HBCD were Oslofjord ≈ Froan > Bear Island >> Spitsbergen (Figure 3B).

Seals. All nine BFR compounds were detected in the harbor seals, except for BDE-209 in seals from the Oslofjord (Table 1). BDE-47 was the most abundant compound in all populations. In the Oslofjord, BDE-99 was the second most abundant compound followed by HBCD. At Froan and Spitsbergen, HBCD was the second most abundant compound, followed by BDE-99 at Froan and BDE-153 at Spitsbergen. Levels of ΣPBDEs (Figure 2C) and HBCD (Figure 3C) differed significantly between the three locations (Kruskal-Wallis, ΣPBDEs: $\chi^2 = 15.47$, df = 2, $p = 0.001$; HBCD: $\chi^2 = 12.86$, df = 2, $p = 0.002$), and were highest in harbor seals from the Oslofjord, somewhat lower in the seals from Froan (Mann-Whitney *U*-test, ΣPBDEs: $n = 14$, $z = -3.000$, $p = 0.001$; HBCD: $n = 14$,

Table 1. Concentrations of BDE congeners, ΣPBDEs, and HBCD in species from different trophic levels in marine coastal ecosystems in the Norwegian North-East Atlantic [ng/g lipid weight; mean ± SD (*n*)].

Location, species	BDE-28	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-209	ΣPBDEs	HBCD
Oslofjord									
Calanus (sp)	1.20 ± 0.32 (3)	35.3 ± 0.80 (3)	26.7 ± 12.2 (3)	6.02 ± 2.30 (3)	2.62 ± 0.78 (3)	1.88 ± 0.91 (3)	0.58 ± 0.29 (3)	74.3 ± 27.9 (3)	4.01 ± 2.82 (3)
Atlantic cod	2.18 ± 1.08 (20)	62.0 ± 30.6 (21)	1.91 ± 0.91 (21)	12.5 ± 5.7 (21)	4.69 ± 9.94 (21)	2.11 ± 1.12 (21)	0.64 ± 0.66 (21)	86.0 ± 41.7 (21)	25.6 ± 13.4 (21)
Harbor seal	1.30 ± 0.19 (5)	246 ± 66 (5)	75.5 ± 24.0 (5)	21.3 ± 5.1 (5)	40.4 ± 16.3 (5)	19.3 ± 11.9 (5)	—	404 ± 101 (5)	50.5 ± 23.8 (5)
Common tern	2.04 ± 0.61 (10)	73.0 ± 18.0 (10)	19.6 ± 2.7 (19)	18.6 ± 3.5 (19)	4.78 ± 0.96 (10)	3.15 ± 0.85 (10)	0.26 ± 0.29 (10)	121 ± 25 (19)	36.4 ± 9.3 (10)
Froan									
Calanus finmarchicus	—	0.80 ± 0.05 (3)	0.21 ± 0.00 (3)	0.14 ± 0.00 (3)	—	—	—	1.15 ± 0.06 (3)	—
Atlantic cod	2.34 ± 1.69 (19)	38.6 ± 31.9 (20)	0.88 ± 0.83 (9)	5.85 ± 5.01 (20)	2.10 ± 2.02 (7)	2.10 ± 1.64 (20)	0.64 ± 0.92 (9)	52.5 ± 40.1 (20)	18.7 ± 10.5 (18)
Harbor seal	0.74 ± 0.29 (9)	88.7 ± 34.2 (9)	8.73 ± 3.78 (9)	3.68 ± 1.30 (9)	4.03 ± 2.43 (9)	0.71 ± 0.80 (9)	0.11 ± 0.07 (6)	106.7 ± 41.2 (9)	22.3 ± 11.9 (9)
Arctic tern	1.36 ± 0.59 (9)	53.9 ± 24.4 (10)	18.0 ± 5.2 (10)	12.1 ± 5.2 (10)	5.04 ± 1.38 (10)	4.67 ± 1.99 (10)	0.22 ± 0.13 (10)	95.4 ± 36.0 (10)	17.0 ± 8.8 (10)
Bear Island									
Polar cod	0.48 ± 0.22 (4)	9.52 ± 1.49 (6)	—	1.98 ± 0.36 (6)	—	0.57 ± 0.11 (6)	—	12.6 ± 1.9 (6)	11.7 ± 7.2 (6)
Spitsbergen									
Calanus glacialis[a]	—	0.08 (1)	0.08 (1)	—	—	—	—	0.16 (1)	—
Polar cod[a,b]	0.10 ± 0.02 (7)	0.88 ± 0.41 (7)	0.19 ± 0.17 (6)	0.18 ± 0.11 (7)	—	0.11 ± 0.05 (5)	0.27 ± 0.08 (5)	1.73 ± 0.80 (7)	1.80 ± 0.58 (7)
Harbor seal	0.18 ± 0.06 (5)	17.8 ± 5.5 (5)	0.98 ± 0.73 (0.73)	0.54 ± 0.19 (5)	1.24 ± 1.42 (5)	0.21 ± 0.19 (4)	0.59 ± 0.54 (5)	21.5 ± 4.7 (5)	3.66 ± 1.54 (5)
Ringed seal[a]	1.35 ± 0.81 (6)	49.3 ± 14.2 (6)	2.33 ± 1.46 (6)	4.69 ± 2.68 (6)	0.68 ± 0.33 (6)	0.69 ± 0.48 (6)	—	59.1 ± 19.7 (6)	19.6 ± 7.6 (6)
Arctic tern	0.38 ± 0.11 (10)	22.0 ± 3.9	7.90 ± 2.49 (10)	4.94 ± 1.06 (10)	2.54 ± 1.47 (10)	2.29 ± 0.97 (10)	0.84 ± 0.89 (10)	40.9 ± 8.4 (10)	4.62 ± 1.44 (10)

—, no data.
[a]Data from Sørmo et al. 2006. [b]Mean values were recalculated based on specimens with levels above the detection limit for BDE-99, BDE-154, BDE-209, and HBCD.

$z = -2.067$, $p = 0.42$) and much lower in harbor seals from Spitsbergen compared with the Oslofjord (Mann-Whitney *U*-test, $n = 10$, $z = -3.6110$, $p = 0.008$ for both ΣPBDEs and HBCD) and Froan (Mann-Whitney *U*-test, $n = 14$, $z = -3.000$, $p = 0.001$ for both ΣPBDEs and HBCD).

In ringed seals from Spitsbergen, all compounds were detected (Table 1). BDE-47 was the most abundant compound, followed by HBCD, BDE-100, and BDE-99. At Spitsbergen, both ΣPBDEs and HBCD were significantly higher in ringed seals compared with harbor seals (Mann-Whitney *U*-test, $n = 11$, $z = -2.739$, $p = 0.004$ for both ΣPBDEs and HBCD).

Terns. In common terns from the Oslofjord and in arctic terns from Froan, BDE-47 was the most abundant compound, followed by HBCD, BDE-99, and BDE-100 (Table 1). Also, in arctic terns from Spitsbergen BDE-47 was the most abundant compound, but in these tern eggs levels of BDE-99 and BDE-100 were somewhat higher than levels of HBCD. Levels of ΣPBDEs (Figure 2D) and HBCD (Figure 3D) differed significantly between the locations (Kruskal-Wallis, $\chi^2 = 17.559$, df = 2, $p < 0.001$; HBCD: $\chi^2 = 22.810$, df = 2, $p < 0.001$). Levels of ΣPBDEs did not differ between tern eggs from the Oslofjord and those from Froan (Mann-Whitney *U*-test, $n = 20$, $z = -1.663$, $p = 0.096$), but levels were significantly lower at Spitsbergen than in the Oslofjord (Mann-Whitney *U*-test, $n = 20$, $z = -3.780$, $p < 0.001$) and at Froan (Mann-Whitney *U*-test, $n = 20$, $z = -3.024$, $p = 0.002$). Levels of HBCD in tern eggs were significantly higher in the Oslofjord than at Froan (Mann-Whitney *U*-test, $n = 20$, $z = -3.250$, $p = 0.001$) and at Spitsbergen (Mann-Whitney *U*-test $n = 20$, $z = -3.780$, $p < 0.001$), and higher at Froan than at Spitsbergen (Mann-Whitney *U*-test $n = 20$, $z = -3.402$, $p = 0.001$).

Biomagnification. The compounds BDE-47, BDE-99, and HBCD were biomagnified from cod to harbor seals at all three locations (Table 2). The BMF of BDE-99 was particularly high in the Oslofjord, whereas the BMF of BDE-47 was particularly high at Spitsbergen. BDE-153 was biomagnified in the Oslofjord and at Froan. Because data on BDE-153 in polar cod from Spitsbergen are lacking, it was not possible to estimate the BMF of this compound at Spitsbergen. BDE-28 was biomagnified only at Spitsbergen, whereas BDE-100 and BDE-154 was biomagnified in the Oslofjord and at Spitsbergen but not at Froan. BDE-209 was biomagnified only at Spitsbergen.

Discussion

In North-East Atlantic coastal ecosystems, levels of BFR compounds generally decreased as a function of increasing latitude (Figures 2 and 3). The obvious reason for this is that the use and leakage of BFRs into the environment is higher in urbanized areas along the Norwegian coast than in the almost unpopulated Spitsbergen. High levels of BFRs have been reported in sewage [see review by Law et al. (2006a)], and the source of BFRs in the southern part of the study area is most likely local discharges from urban sewage and industrial activity. Because of their semivolatile properties, POPs are subject to long-range atmospheric transport (Wania and Mackay 1993, 1996), and this is thus most likely the origin of the BFRs detected in endemic Arctic biota.

The finding herein—that levels of BFRs are lower in marine Arctic ecosystems than in temperate marine ecosystems—is also in accordance with previous reports in marine mammals. Data compiled on PBDEs in marine mammals from temperate environments and from the Canadian Arctic showed that levels of PBDEs were about 1,000 times higher in marine mammals from temperate marine ecosystems than in ringed seals from the Arctic (Ikonomou et al. 2002). Furthermore, levels of ΣPBDEs (BDE-17, BDE-47, BDE-49, BDE-99, BDE-100, BDE-119, BDE-140, BDE-153, BDE-154, BDE-183) in harbor porpoises also decreased as a function of increasing latitude along the Norwegian coast, and were lower in animals from Iceland than from Norway (Thron et al. 2004). Concentrations of polychlorinated biphenyls (PCBs) in seawater in the North-East Atlantic have been shown to decrease as function of increasing latitude (Sobek and Gustafsson 2004). This confirms

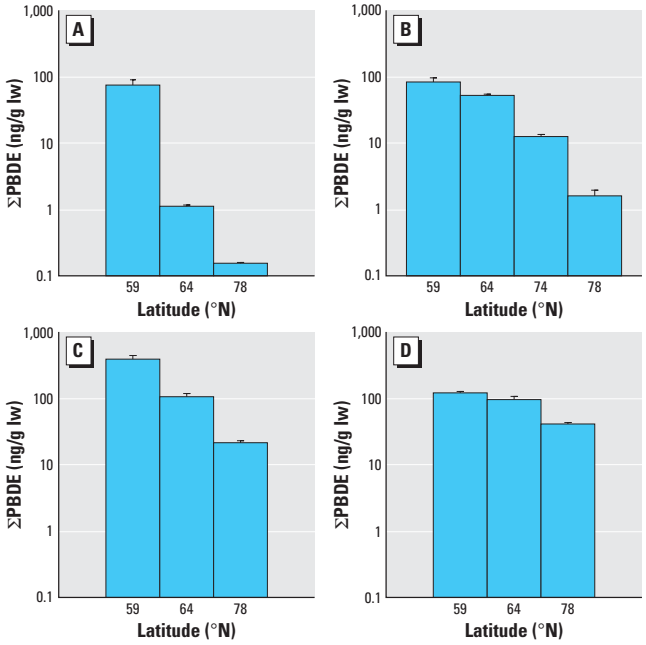

Figure 2. Concentrations of ΣPBDEs (ng/g lw ± SE) in (*A*) calanoids, (*B*) cod [Atlantic cod (*Gadus morhua*) at 59° N and 64° N, and polar cod (*Boreogadus saida*) at 74° N and > 78° N], (*C*) harbor seal (*Phoca vitulina*), and (*D*) terns [common tern (*Sterna hirundo*) at 59° N; arctic tern (*Sterna paradisea*) at 64° N and 78° N].

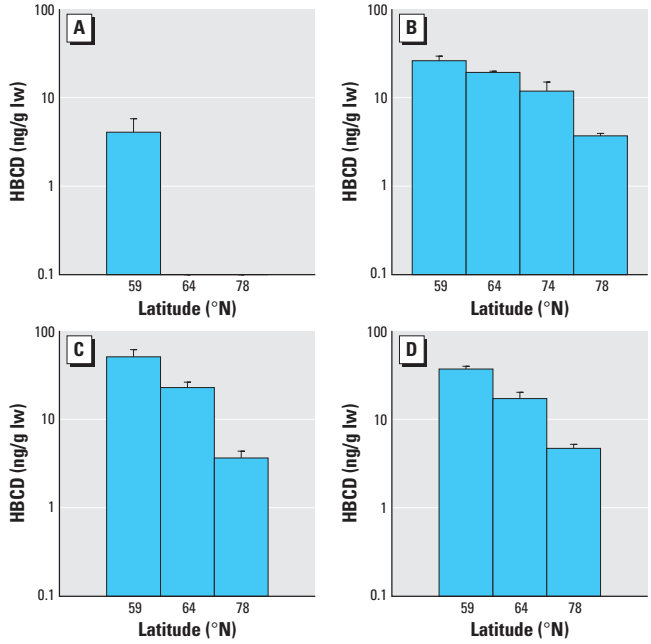

Figure 3. Concentrations of HBCD (ng/g lw ± SE) in (*A*) calanoids, (*B*) cod [Atlantic cod (*Gadus morhua*) at 59° N and 64° N, and polar cod (*Boreogadus saida*) at 74° N and > 78° N], (*C*) harbor seal (*Phoca vitulina*), and (*D*) terns [common tern (*Sterna hirundo*) at 59° N; arctic tern (*Sterna paradisea*) at 64° N and 78° N].

that levels of POPs in marine biota generally decrease as a function of the distance from the release areas.

When we compare organisms that occupy similar trophic levels, the Arctic is still a pristine environment with respect to organohalogenated anthropogenic compounds. This is contrary to the beliefs of many politicians, governmental bureaucrats, and nongovernmental organizations, who, because of the particularly high levels of PCBs reported in polar bears (*Ursus maritimus*), seem to believe that the Arctic is heavily polluted by POPs. The high levels of PCBs in polar bears are attributed to the fact that this species is an apex predator that feeds almost exclusively on the blubber of seals (Derocher et al. 2002). Thus, because of biomagnification of the most persistent PCB congeners from seals to polar bears, levels of ΣPCB in polar bears become very high (Bernhoft et al. 1997; Skaare et al. 2002). Levels of all BFR compounds analyzed herein (except for BDE-153) were lower in polar bears than in its main prey species, the ringed seal (Sørmo et al. 2006), most likely because the polar bear has a high ability to metabolize POPs (Letcher et al. 1996).

The clear latitudinal decrease in levels of BFRs was not that pronounced in the two tern species compared with the other species included in the study (Figures 2 and 3). This is most likely linked to the fact that the terns are migratory birds, whereas the other species are endemic to their regions. During their migration from Africa (common tern) and Antarctica (arctic tern), they feed along the highly urbanized and thus more polluted coasts of Europe. Therefore, even though the terns may metabolize and excrete some of the BFR compounds during their migration via urbanized and industrialized polluted areas, levels still seem to be relatively high when they reach their breeding areas. Because migration is energetically costly, the birds will have to build up lipid stores for egg laying when arriving at their breeding sites. Thus, because levels of POPs are lower in prey at more northern breeding sites, body burdens of POPs in female birds will be diluted, resulting in the latitudinal decrease of BFR compounds in their eggs reported herein. When the eggs are laid, the lipophilic BFRs are transferred from the female to her eggs.

Table 2. Biomagnification factors from Atlantic cod (*Gadus morhua*) to harbor seal (*Phoca vitulina*) (Oslofjord and Froan), and from polar cod (*Boreogadus saida*) to harbor seal (Spitsbergen).

Compound	Oslofjord	Froan	Spitsbergen
BDE-28	0.6	0.3	1.8
BDE-47	4.0	2.3	20.2
BDE-99	39.5	9.9	5.2
BDE-100	1.7	0.6	3.0
BDE-153	8.6	1.9	
BDE-154	9.1	0.3	1.9
BDE-209		0.2	2.2
ΣPBDE	4.7	2.0	12.4
HBCD	2.0	1.2	2.0

The high levels of HBCD reported in common tern eggs from the Netherlands [330–7,100 ng/g lw (Morris et al. 2004)] occur most likely because specimens that breed here are exposed to higher concentrations for a longer period of time than specimens that only transiently pass the Netherlands en route to Norway and the Arctic.

In another study on kittiwakes (*Rissa tridactyla*), levels of the sum of 23 PCB congeners and the ΣPBDEs (BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154) in newly hatched chicks did not differ between the west coast of Norway (Runde, 62° N) and Spitsbergen (Kongsfjorden, 79° N) (Murvoll et al. 2006b). The apparent lack of a latitudinal decrease in PBDE levels in kittiwakes may be because they winter in the North Atlantic, close to where these compounds are used and released.

In calanoid species, HBCD was detected only in the Oslofjord (Figure 3A). In Atlantic cod, levels of PBDEs (Figure 2B) and HBCD (Figure 3B) were somewhat higher in the Oslofjord than at Froan, and were lowest in polar cod from Spitsbergen. Levels of PBDEs in the Oslofjord were considerably lower than concentrations reported in cod from 16 different locations in the North Sea and Skagerrak (Boon et al. 2002).

Because food webs are complex, and because few species were studied, we acknowledge that it is difficult to estimate biomagnification rates in the different ecosystems included in this study. However, our crude approach, assuming a simple cod–harbor seal food chain, will still give some information on biomagnification processes of BFR compounds in the three coastal ecosystems.

In the Oslofjord, the biomagnification of BDE-99 from Atlantic cod to harbor seal was particularly high (Table 2), perhaps because BDE-99 constitutes a large part of the technical penta-BDE mixture, and the releases of this compound into the environment probably has been high. The high level of BDE-99 reported in the nearby Drammens fjord (Zegers et al. 2003) supports this. Furthermore, levels of BDE-99 were quite high in calanoids from the Oslofjord (Table 1). Much higher levels of BDE-99 than reported herein have been reported in more pelagic stocks of Atlantic cod in the Skagerrak and the North Sea (Boon et al. 2002). It is also possible that harbor seals in the Oslofjord prefer to prey on cod from the pelagic stocks. The Atlantic cod sampled in this study may have belonged to a more coastal bound stock which the harbor seal does not prefer to prey on, and this may have resulted in an overestimation of the BMF for BDE-99. It should also be noted that BDE-99 is *meta-para*-substituted and consequently not easily metabolized (Veltman et al. 2005). These factors may help explain the high BMF of BDE-99 from Atlantic cod to harbor seals in the Oslofjord.

The BMF of BDE-153 was also high in harbor seals from the Oslofjord, perhaps because BDE-153 has a substitution pattern similar to that of PCB-153, which is the most persistent PCB compound. It is therefore possible that the high BMF of BDE-153 in the Oslofjord is caused by its persistence.

In most species from all locations, BDE-47 was the most abundant congener (Table 1). This congener was also biomagnified throughout the three food chains (Table 2). BDE-47 constitutes approximately 25% of the technical penta-BDE mixture (Hites 2004) and has thus been released to the environment in relatively large volumes. Furthermore, there are indications that in fish, BDE-99 (which constitutes ~ 50% of the penta-BDE mixture) is debrominated to BDE-47 (Stapleton et al. 2004), and this may thus lead to a further biomagnification of BDE-47 in marine food chains.

Even though BDE-209 often is the predominating PBDE congener in marine sediments (de Boer et al. 2003), it has been reported to contribute very little to the total PBDE burden in organisms (Law et al. 2006a). This is believed to be caused by the large molecular size of the compound and the resultant low transfer over cells and uptake into the organisms (Stapleton et al. 2004). Recently, there has been a growing body of evidence that suggests that BDE-209 is bioaccumulated to a larger extent in terrestrial food chains than in marine food chains (Law et al. 2006a). However, BDE-209 has been reported to account for > 50% of total BDE burden in the detritus feeding ice-amphipod *Gamarus wilkitzkii* at Spitsbergen (Sørmo et al. 2006). Herein, BDE-209 was detected in animals from all the three ecosystems (Table 1). Because BDE-209 is almost ubiquitous, all possible efforts were made to avoid contamination of the samples during sampling, storage, and analysis. During the analyses, blank samples were run parallel to the samples to control for possible contamination in the laboratory, and no such contamination could be identified.

The highest BDE-209 levels were found in arctic tern eggs from Spitsbergen (Table 1). Further, it should be noted that the highest concentration of BDE-209 relative to ΣPBDEs was found in polar cod from Spitsbergen (ca. 16% of ΣPBDEs), harbor seals from Spitsbergen (~ 3%), and arctic terns from Spitsbergen (~ 2%).

BDE-209 has a strong affinity to particles. It is therefore possible that the detected levels in the calanoid species and in the two cod species are associated with the cuticle/skin or

sediment particles and/or prey species in the intestines (Law et al. 2006a; Leonards et al. 2004). However, the detection of BDE-209 in tern eggs and seal blubber shows that it is accumulated also in marine food chains. This is consistent with reports that BDE-209 was bioaccumulated in grey seal given a supplement of this congener in their diet (Thomas et al. 2005). BDE-209 has recently also been reported in adipose tissue and plasma from polar bears and glaucous gulls (*Larus hyperboreus*) from Spitsbergen (Sørmo et al. 2006; Verreault et al. 2005). The relatively high contribution of BDE-209 to ΣPBDEs in animals from Spitsbergen demonstrates that this congener is subject to long-range transport and dispersal.

Whereas the data herein indicate that BDE-209 may be biomagnified from polar cod to harbor seals (Table 2), this was not the case from Atlantic cod to seals at Froan. Thus, the potential of BDE-209 to be transferred in food webs is unclear. The technical deca-BDE-mixture, in which BDE-209 is the major congener, presently constitutes about 80% of the world market demand of PBDEs (de Boer et al. 2003). Thus, there is a clear need for more information on the ability of BDE-209 to biomagnify and/or be debrominated in marine ecosystems.

At Spitsbergen, levels of both PBDEs and HBCD were higher in ringed seals than in harbor seals (Table 1). The most obvious differences between these two seal species were related to the much higher levels of BDE-47, HBCD, and BDE-100 in ringed seals. These differences are most likely related to differences in species-specific differences in their ability to metabolize and biotransform the BFR compounds, and possibly also related to differences in prey preferences.

There are few reports concerning levels of HBCD in marine ecosystems (Morris et al. 2004; Stapleton et al. 2006; Wolkers et al. 2004; Zegers et al. 2005). Herein, HBCD were found in animals from all trophic levels, except in calanoids at Froan and at Spitsbergen. The commercial HBCD mixtures mainly consist of the three stereoisomers γ-HBCD (75–89%), α-HBCD (10–13%), and β-HBCD (1–12%) (Heeb et al. 2005). In biota, the HBCD isomer composition changes, and α-HBCD dominates (Law et al. 2006a). In the present study, we did not distinguish among the different isomers of HBCD. However, in aquatic invertebrates, marine fish, birds, and marine mammals HBCD is present predominantly as α-HBCD (Covaci et al. 2006).

In cod, seals, and terns, HBCD levels seemed to be similar in the Oslofjord and at Froan, whereas levels were much lower at Spitsbergen (Figure 3B–D), except in ringed seals (Table 1). The particular high levels of HBCD in the ringed seals indicate that the bioaccumulation potential of HBCD in this species may be particularly high (Sørmo et al. 2006). Previously, it has been reported that HBCD does not seem to biomagnify from ringed seals to polar bears (Sørmo et al. 2006) possibly because polar bears generally have a large capacity to metabolize organohalogenated compounds (Letcher et al. 1996).

In common dolphins (*Delphinus delphis*) from the Central and South Atlantic coast of Europe (Scotland, Ireland, the Netherlands, Spain), median concentrations of HBCD ranged from 200 to 900 ng/g lw, whereas median concentrations in harbor porpoises ranged from 100 to 5,100 ng/g lw (Zegers et al. 2005). Levels were highest in the Irish Sea and in North-East Scotland. In blubber of two harbor seals from the western Wadden Sea, concentrations of HBCD ranged from 63 to 2,055 ng/g lw (Morris et al. 2004). In stranded and by-caught harbor porpoises in the United Kingdom, HBCD levels ranged from 11 to 21,300 ng/g lw (Law et al. 2006b), and a time-trend analysis of the data strongly indicated a sharp increase in HBCD concentrations from about 2001 onward. The HBCD levels reported in cetaceans from the South- and Central-East Atlantic coast, and in the harbor seals from the Wadden Sea are much higher than those found in harbor seals from the Oslofjord, Froan, and Spitsbergen (Table 1).

Relatively high levels of HBCD have also been reported in hatchlings of kittiwakes from the Norwegian west coast (~ 260 ng/g lw; Runde, 62° N) and levels were somewhat lower in hatchlings from Spitsbergen (~ 120 ng/g lw; Kongsfjorden 79° N) (Murvoll et al. 2006b). Furthermore, even higher levels of HBCD were reported in hatchlings of European shags (*Phalacrocorax aristotelis*) from the western Norwegian coast (~ 420 ng/g lw; Sklinna 65° N) (Murvoll et al. 2006a). The much higher levels in the European shag and kittiwake hatchlings may be related to differences in the analytical matrix (whole egg herein vs. yolk sac in hatchlings). However, the differences may also be related to the fact that kittiwakes and European shags winter in the North Sea, the Norwegian Sea, and along the Canadian east coast, whereas common and arctic terns migrate to the more pristine areas in Africa and Antarctica, respectively. In North Sea estuaries (United Kingdom and the Netherlands), levels of HBCD in cormorant livers (*Phalacrocorax carbo*) and common tern eggs were 330–7,100 and 138–1,320 ng/g lw, respectively (Morris et al. 2004). These concentrations are higher than those reported in the tern eggs herein.

Because HBCD has been reported to have histopathologic and neurotoxic effects (Birnbaum and Staskal 2004; Darnerud 2003; Mariussen and Fonnum 2003), there is cause for concern about the spreading and uptake of this compound in biota. In Californian sea lions (*Zalophus californianus*) a significant temporal increase in HBCD was reported from 1994 to 2004 (Stapleton et al. 2006), and in harbor porpoises from the United Kingdom a sharp increase in HBCD concentrations was found from about 2001 onward (Law et al. 2006b). Because there currently are no restrictions on the use of HBCD (Stapleton et al. 2006), there are reasons to believe that the global spreading of the compound will continue and that levels in Arctic biota will increase with time.

Conclusions

Levels of BFRs in Arctic North-East Atlantic coastal ecosystems (Spitsbergen) are generally lower than along the Norwegian coast and much lower than in South- and Central-East Atlantic coastal ecosystems. This reflects the distance from the release sources. The identification of BDE-209 and HBCD in animals from all trophic levels and the relatively high contribution of BDE-209 to ΣPBDEs in some Arctic animals warrant the need for further focus on the global spreading and biomagnification potential of these compounds, because they are currently in unrestricted use.

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